Z. Phys. B – Condensed Matter 77, 95–104 (1989)

Condensed Zeitschrift für Physik B Matter

Magnetic order in a Kondo lattice: a neutron scattering study of $CeCu_2Ge_2$

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Received July 6, 1988; revised version June 3, 1989

Elastic and inelastic neutron scattering studies of the Kondo lattice $CeCu_2Ge_2$ were performed. At $T_N = 4.1$ K an incommensurate magnetic order develops with an ordering wave vector $\mathbf{q}_0 = (0.28, 0.28, 0.54)$ and an ordered moment $\mu_s = 0.74\ \mu_B$. The crystalline electric field splits the $4f^1$-J-multiplet of the Ce ion into a ground state doublet and a quartet at 191 K. The wave function of the ground state yields an ordered moment of $1.54\ \mu_B$. Thus, due to the onset of the formation of a Kondo singlet the magnetic moment is considerably reduced. The magnetic relaxation rate Γ was investigated via quasielastic neutron scattering. The temperature dependence of $\Gamma(T)$ is characteristic of heavy-fermion systems with a high temperature square root dependence and a limiting low temperature value, yielding a Kondo temperature $T_K \approx 10$ K. The quasielastic component of the scattered neutron intensities persists down to the lowest temperatures, well below T_N. This quasielastic line is regarded as a characteristic feature of heavy-fermion systems and corresponds to the enhanced value of the linear term of the specific heat.

I. Introduction

During the last decade a new branch of material science has developed dealing with stoichiometric systems with contain Ce- or U-ions and which exhibit very unusual low temperature properties [1]: A very large linear term of the specific heat and an enhanced, almost temperature independent Pauli spin susceptibility correspond to high effective masses of the electronic quasiparticles. Consequently these compounds have been termed Heavy-Fermion Systems (HFS). The ground state of HFS can be quite different: the onset of superconductivity has been reported for $CeCu_2Si_2$ [2], UBe_{13} [3] and UPt_3 [4]. U_2Zn_{17} [5], UCd_{11} [6] and $CeCu_2Ge_2$ [7, 8] show antiferromagnetism at low temperatures and a pure heavy-fermion state has been found to exist in $CeAl_3$ [9] and $CeCu_6$ [10]. These nonmagnetic and non-superconducting HFS exhibit a coherent state at $T < T_{coh} \ll T_K$, where T_K is the usual Kondo temperature (in this paper T_K denotes the characteristic energy scale of the Kondo lattice). In this regime coherent scattering of the conduction electrons by the lattice of the Kondo ions is the dominant interaction. As a consequence a fine structure develops in the electronic density of states at the Fermi level. Experimentally coherence effects are sometimes even reflected in the thermodynamic properties: e.g. for $CeAl_3$ and $CeCu_2Si_2$ a maximum appears in C/T at T_{coh} followed by a decrease of C/T for further decreasing temperatures [11].

$CeCu_2Ge_2$ is an isostructural compound to the prototypical HFS $CeCu_2Si_2$. The resistivity versus temperature curves in $CeCu_2Ge_2$ [12] closely resemble those measured for $CeCu_2Si_2$ [13] in that they exhibit two maxima in $\rho(T)$. The maximum at higher temperatures is due to crystal electric field effects, the second maximum at $T \approx 6$ K is probably due to the transition into a heavy-fermion state, indicating a

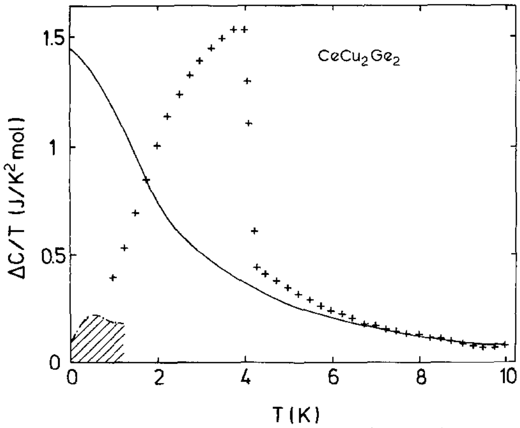

Fig. 1. Magnetic contribution ΔC to the specific heat plotted as $\Delta C/T$ versus T in $CeCu_2Ge_2$. The low temperature data (shaded area) were obtained by subtracting nuclear and magnon contributions from the raw data [7]. The solid line is the result of a calculation [15] using a Bethe-Ansatz for a spin 1/2 Kondo impurity model with a Kondo temperature $T_K = 6$ K

Kondo temperature $T_K \approx 6$ K. At $T = 4$ K a sharp drop in the resistivity reflects the formation of a magnetically ordered state. A Kondo temperature of 6 K is roughly consistent with the results of thermoelectric power experiments [14] with a low temperature minimum at $T \approx 15$ K. Phenomenologically the position of the minimum is located at $2T_K$. However, one has to stress that these estimates of the Kondo temperature from resistivity and from thermoelectric power data have no theoretical basis (In a recent theoretical work by Cox and Grewe [15] it has been shown that the electric resistivity peaks just below the characteristic Kondo scale). The most unexpected result for $CeCu_2Ge_2$ was the enhanced linear term of the specific heat at temperatures well below the magnetic phase transition [7]. In addition, for $T \leqq 0.5$ K the C/T versus T data indicate the importance of coherence effects [7] as discussed in the case of $CeAl_3$ and $CeCuSi_2$ [11]. In Fig. 1 we have replotted the experimental results of $\Delta C/T$ from [7] showing the non-phononic contribution to the specific heat, only. The dash-dotted line for $T \leqq 1.2$ K gives the electronic specific heat after subtracting both, the nuclear and the magnon contributions [7]. The solid line represents the result of the specific heat using a Bethe-Ansatz for a spin-1/2 Kondo impurity model as derived by Desgranges and Schotte [15] using a Kondo temperature $T_K = 6$ K. The T-dependence of the specific heat for $T \geqq 4$ K is well reproduced and this model yields $\gamma(T=0) = 1400$ mJ/K^2-mol, which characterizes $CeCu_2Ge_2$ as a HFS. Of course, the onset of magnetic order at $T = 4$ K suppresses the further development of the heavy-fermion state.

The experiments performed so far characterize $CeCu_2Ge_2$ as a magnetically ordered Kondo lattice with a Kondo temperature $T_K = 6$ K and a magnetic phase transition $T_N = 4.1$ K. Despite the presence of long range magnetic order, the aforementioned coherence effects, which are typical of non-magnetic HFS, still seem to play an important role. The aim of the present elastic and inelastic neutron scattering experiments was to determine *i)* the magnetic structure and the size of the ordered moment, *ii)* the ground state and the splitting of the $4f^1$ J-multiplet in the presence of the crystalline electric field (CEF) and *iii)* the temperature dependence of the magnetic relaxation rate. The results regarding points *i)* and *ii)* should allow a determination of the reduction of the magnetic moment of the CEF ground state due to the Kondo effect. The magnetic relaxation rate at $T \to 0$ yields a reliable estimate of the Kondo temperature. In addition, it appears worthwhile to investigate the line width and the energy of the magnetic excitations in the magnetically ordered phase. In $Zn_{17}U_2$, another Heavy Fermion System with a magnetic ground state, no sharp magnetic excitations at non-zero energies could be detected in the antiferromagnetic state [17].

II. Experimental results and analysis

Polycrystalline samples of $CeCu_2Ge_2$ have been melted from stoichiometric amounts of high purity elements in an argon-arc furnace. At room temperature the samples were investigated by X-ray powder diffraction techniques. The samples exhibited the proper $ThCr_2Si_2$ structure with lattice parameters $a = 4.17$ Å and $c = 10.21$ Å.

A. Neutron diffraction experiments

In order to investigate the structure and the size of the ordered magnetic moment, neutron powder diffraction experiments have been performed utilizing the multidetector diffractometer D 1 B located on a thermal neutron guide at the high flux reactor at the Institut Laue-Langevin in Grenoble. The wave length of the incident neutrons was 2.52 Å. A powder diffraction spectrum recorded at 1.5 K is shown in Fig. 2 (upper part). The sample exhibits predominantly a single phase $ThCr_2Si_2$ structure. However, small amounts of spurious phases could be detected (shaded areas in the upper part of Fig. 2). The lattice constants of the nuclear structure as determined at low temperatures are: $a = 4.158$ Å, $c = 10.227$ Å and $z = 0.385$ Å (z is the reduced coordinate of the Ge atoms in the tetragonal cell and is determined from the intensities of the nuclear Bragg reflections). The lower part of Fig. 1 shows the difference spectrum: here the intensi-

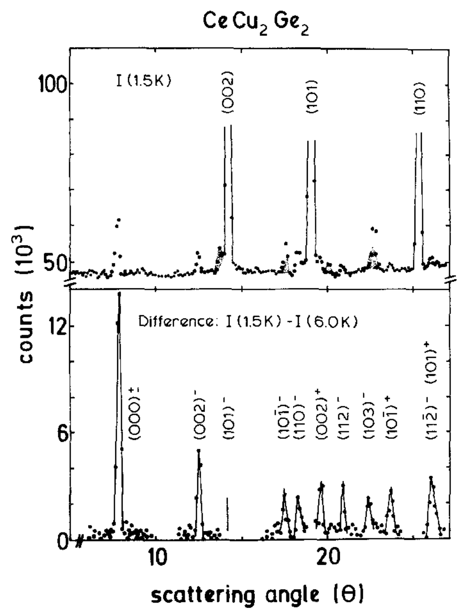

Fig. 2. *Upper Frame:* Raw data of nuclear and magnetic Bragg reflections in $CeCu_2Ge_2$ at 1.5 K. The shaded areas are nuclear reflections due to parasitic phases. *Lower Frame:* Difference spectra in $CeCu_2Ge_2$ (I(1.5 K)$-I$(6 K)) yielding the magnetic Bragg reflections, only. The (1 0 1)$^-$ reflection was screened by the (0 0 2) nuclear Bragg reflection

ties as measured in the paramagnetic phase at 6 K were subtracted from the spectrum as observed in the magnetically ordered phase at 1.5 K. Even from a rough inspection of the magnetic diffraction pattern it is obvious that the magnetic structure exhibits neither a ferromagnetic nor a simple type of antiferromagnetic arrangement. The Bragg angles of the magnetic reflections can be indexed in terms of $|\mathbf{Q}| = |\tau_{hkl} \pm \mathbf{q}_0| \equiv (hkl)^{\pm}$ where τ_{hkl} is a vector of the reciprocal chemical lattice and $\mathbf{q}_0$ is the propagation vector of a modulated spin arrangement. $\mathbf{q}_0$ has been determined by trial and error and a best fit to the experimental data has been achieved with $\mathbf{q}_0 = (0.28, 0.28, 0.54)$, in reduced units $2\pi/a$, $2\pi/a$, $2\pi/c$, respectively. Two models of modulated spin structures have been examined: spirals, including conical spirals, and collinear arrangements with a modulation of the length of the magnetic moment (usually referred to as longitudinal static spin waves). Better fits to the observed intensities have been obtained with spirals. The structure factor of the satellite reflections of the magnetic spiral structure is given by

$$F_{hkl} \propto \frac{\sqrt{1+\cos^2 \eta}}{2} \sum_{v} f_v \sin \beta_v \, e^{i\phi_v} \cdot \exp[2\pi i(hx_v + ky_v + lz_v)], \qquad (1)$$

where x_v, y_v, z_v are the reduced coordinates of the two magnetic ions with reference to the chemical cell,

Table 1. Calculated and observed magnetic structure factors and peak positions in $CeCu_2Ge_2$ using $a = 4.158$ Å and $c = 10.227$ Å and a propagation vector $\mathbf{q}_0 = (0.28, 0.28, 0.54)$. The model used was a magnetic spiral with a plane of rotation perpendicular to the propagation vector $\mathbf{q}_0$ and a magnetic moment $\mu_S = 0.74 \mu_B$. The $(1\,0\,1)^-$ reflection could not be determined experimentally since it is screened by the strong (002) nuclear Bragg peak

$(h\ k\ l)$	Θ_{calc}	Θ_{obs}	F^2_{calc}	F^2_{obs}
$(0\ 0\ 0)^{\pm}$	7.81	7.88	1.59	1.48
$(0\ 0\ 2)^-$	12.47	12.49	1.13	1.21
$(1\ 0\ 1)^-$	13.92	–	2.44	–
$(1\ 0\ \bar{1})^-$	17.51	17.45	2.24	1.35
$(1\ 1\ 0)^-$	18.38	18.29	1.40	1.28
$(0\ 0\ 2)^+$	19.56	19.61	1.41	1.80
$(1\ 1\ 2)^-$	20.90	20.86	1.58	1.81
$(1\ 0\ 3)^-$	22.49	22.55	–	–
$(1\ 0\ 1)^+$	23.61	23.65	2.68	2.50
$(1\ 1\ 2)^-$	26.04		1.15	
		26.11		4.70
$(1\ 0\ 1)^+$	26.07		2.97	

f_v is the magnetic form factor of Ce^{3+}, η is the angle between the axis of the spiral and the scattering vector $\mathbf{Q}$, β_v is the semiaperture angle of the spiral, and $e^{i\phi_v}$ allows for the phase relations between different spirals. Since there are no magnetic contributions to the chemical Bragg peaks, $\beta_v = 90$ deg. The two Ce ions of the tetragonal cell are equivalent. Hence the two phase angles are identical to $\phi_1 = \phi_2$ and they can be set to zero. The magnetic structure reduces to a single plane spiral. The best fit was obtained with a plane of rotation perpendicular to the propagation vector $\mathbf{q}_0$ and a moment $\mu_s = 0.74\ \mu_B$ at 1.5 K. In direct space the spiral propagates almost along the body diagonal of the tetragonal lattice which is the bond direction of two neighboring Ce ions. The deviation from the body diagonal is 6 deg. In Table 1 we show the calculated and the observed values of the peak positions and of the magnetic structure factors. Figure 3 shows the temperature dependence of the square root of the intensity of the first magnetic reflection, $(0\,0\,0)^{\pm}$. It can be regarded as a linear measure of the ordered moment. Variations of the propagation vector $\mathbf{q}_0$ with temperature have not been observed.

B. Inelastic neutron scattering experiments

Inelastic neutron scattering experiments were performed in order to study the crystal electric field splitting and the temperature dependence of the magnetic relaxation rate in $CeCu_2Ge_2$. The latter is determined by the line width of transitions within the ground state of the $4f^1$-multiplet.

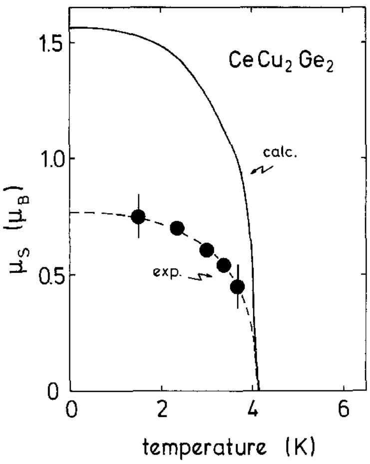

Fig. 3. Temperature dependence of the ordered magnetic moment as determined from the peak intensities of the $(0\,0\,0)^{\pm}$ reflection, rescaled to the averaged magnetic moment of $0.74\mu_B$ at 1.5 K. The solid line is the result of a mean field calculation of μ_S utilizing the appropriate ground state of Ce^{3+} ion in the crystal electric field and an ordering temperature of 4.1 K

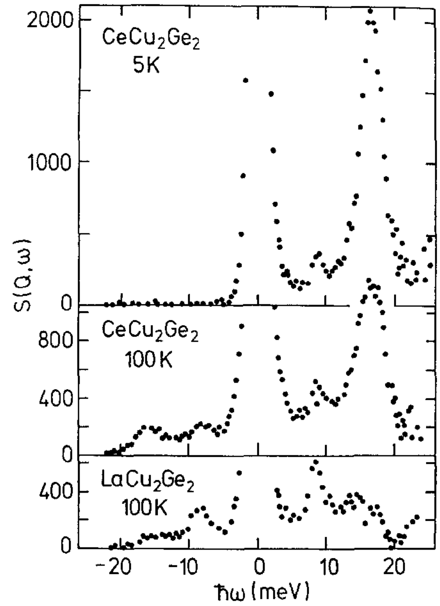

Fig. 4. Scattering law for an average scattering angle $\Theta = 11.3°$ as a function of neutron energy transfer $\hbar\omega$ as obtained for $CeCu_2Ge_2$ at 5 K and 100 K and for $LaCu_2Ge_2$ at 100 K

The double differential cross section for magnetic scattering is proportional to the scattering law $S(Q, \omega)$ which in turn is related to the imaginary part of the wave number and frequency dependent magnetic susceptibility $\chi(Q, \omega)$ via

$$\frac{d^2\sigma}{d\Omega\, d\omega} \propto S(Q, \omega) = \left(1 - \exp\frac{\hbar\omega}{k_B T}\right)^{-1} \mathrm{Im}\,[\chi(\mathbf{Q}, \omega)]. \quad (2)$$

For an ensemble of N non-interacting Ce ions the imaginary part of $\chi(\mathbf{Q}, \omega)$ consists of a sequence of Lorentzian lines defining energies and lifetime effects of the CEF eigenfunctions. Using the dipole approximation for small Q-values and ignoring lifetime effects for the moment, the cross section is given by

$$\frac{d^2\sigma}{d\Omega\, d\omega} = N \cdot \rho^2 \cdot \frac{k_f}{k_i} f^2(\mathbf{Q}) \sum_{ij} \eta_i \langle j | J_\perp | i \rangle^2 \cdot \delta(E_j - E_i - \hbar\omega). \quad (3)$$

Here k_i and k_f are the wave numbers of incoming and outgoing neutrons, $\mathbf{Q}$ and $\hbar\omega$ are the momentum and energy transfer, respectively. The CEF states $|i\rangle$ have energies E_i and thermal occupation probabilities n_i. $f(\mathbf{Q})$ is the magnetic form factor and $\rho = 0.0724$ barn/sr. $J_\perp$ is the component of the total angular momentum $\mathbf{J}$ perpendicular to $\mathbf{Q}$. Here we have neglected the Debye-Waller factor and assumed that life time effects are negligible. This formalism accounts for both elastic and inelastic scattering processes, where $i = j$ describes Curie-type elastic processes and $i \neq j$ Van Vleck-type of transitions. In the neutron scattering experiments of the present investigation we utilized thermal neutrons to study inelastic transitions. Cold neutrons were used to investigate life time effects of the elastic processes within the ground state of the CEF level scheme.

1. Crystal electric field splitting. Inelastic neutron scattering experiments in a range of neutron energy transfers -50 meV $\leqq \hbar\omega \leqq 50$ meV were performed to determine the splitting of the $4f^1$ ground state J-multiplet in the presence of the CEF. The magnetic moment of the ground state and the crystal electric field splitting are important parameters for any detailed analysis of the properties of Kondo lattices. These experiments were carried out using the time-of-flight spectrometer IN4 located on a thermal neutron source at the Institute Laue Langevin. Incident neutron energies of $E_0 = 12.5$, 30 and 50 meV were utilized. Figure 4 shows the neutron spectra of $CeCu_2Ge_2$ and $LaCu_2Ge_2$ as measured with an incident neutron energy of 30 meV and taken at an average scattering angle of 11.3 deg. An identification of the magnetic component of the spectra is possible by comparing spectra at different temperatures and at different scattering angles. In addition, the neutron spectra of the nonmagnetic isostructural compound $LaCu_2Ge_2$ allows an identification of phonon scattering contributions. Figure 4 suggests that the excitation at $\hbar\omega = 16$ meV is of magnetic, whereas the exci-

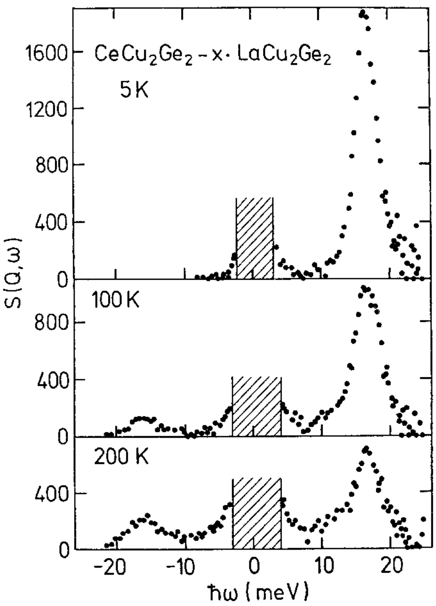

Fig. 5. The difference spectra $CeCu_2Ge_2 - x\,LaCu_2Ge_2$ at 5 K, 100 K and 200 K. The factor x is estimated from the difference of the nuclear coherent scattering lengths of $CeCu_2Ge_2$ and $LaCu_2Ge_2$ and the different phonon populations at 5 K, 100 K and 200 K

tation at 8 meV is of phononic origin. Considering the different nuclear coherent scattering lengths and masses of La and Ce we arrived at estimates for non-magnetic contributions to the scattered intensities and corrected the spectra correspondingly. Corrected spectra at 5 K, 100 K and 200 K are shown in Fig. 5. At 5 K a single peak is visible only at the neutron energy loss side of the spectrum. With increasing temperature, due to the population of the higher crystal electric field levels, the reverse excitation line appears on the neutron energy gain side. The measurements with the lower and with the higher incident neutron energies did not give any evidence for further CEF transitions within the range of energy transfers, $1\ \text{meV} \leqq \hbar\omega \leqq 50\ \text{meV}$, accessible in present time of-flight experiments. For $T \gtrsim 100$ K one notes some feeble extra intensity in the $\hbar\omega$-range from 9 to 13 meV. The Q- and T-dependence suggest that it is of phononic origin. Obviously, this feature is not corrected properly by calculating the difference spectra. Thus the neutron experiment observes a single CEF transition. Since the spectroscopic ground state of the Ce-ion is split into three doublets (see below), two types of CEF level schemes are consistent with the neutron results: three about equally spaced doublets, or an effective two level system, doublet-quartet. Only the second choice, with a doublet ground state, is in agreement with specific heat measurements. It will be considered in the following.

In the present compound the Ce sites are equivalent and have tetragonal point symmetry. The CEF-Hamiltonian is of the form

$$H_{\text{CEF}} = B_2^0\, O_2^0 + B_4^0\, O_4^0 + B_4^4\, O_4^4.$$

Here the fourfold c-axis is the axis of quantization. The CEF-parameters B_n^m and the operator equivalents O_m^n have been defined by Hutchings [19]. The Hamiltonian acts on the $J = 5/2$-multiplet of Ce^{3+} which is split into three doublets D_i:

$$D_1 = |\pm 1/2\rangle$$
$$D_2 = a|3/2\rangle + b|-5/2\rangle, \quad +b|5/2\rangle + a|-3/2\rangle$$
$$D_3 = a|5/2\rangle - b|-3/2\rangle, \quad -b|3/2\rangle + a|-5/2\rangle$$
with $a^2 + b^2 = 1$.

For $B_4^4 = 0$, $a = 1$, and $b = 0$. The three doublets are mutually connected by magnetic dipole transitions. The $D_1 - D_2$-transition matrix element vanishes for $B_4^4 = 0$. Thus, in the general case three transitions can be observed in an inelastic neutron scattering experiment. In the present case, however, where two of the three doublets are degenerate, the information from this type of experiment is insufficient for the determination of the three CEF-parameters.

We arrive at a determination of the CEF parameters through the following reasoning. The measurements of the paramagnetic low field (4 mT) susceptibility [7] rules out $a|\pm 1/2\rangle$ ground state, since in this situation distinct deviations from a Curie-Weiss behavior should have been observed. The energies of the two excited doublets Δ_1 and Δ_2 with $\Delta_1 = \Delta_2$, one of them being the $|\pm 1/2\rangle$ state, supply two pieces of information for the determination of the three parameters B_2^0, B_4^0 and B_4^4. Hence the solution is one point on a one-dimensional manifold in the three-dimensional parameter space. We have determined this manifold by calculating B_4^0 and B_4^4 from the values of Δ_1 and Δ_2 determined from the time-of-flight experiments for a given value of B_2^0 and then varied B_2^0. Simultaneously, the paramagnetic susceptibility $\chi = \frac{M}{H}$ and the magnetization M have been calculated for each set of B_2^0, B_4^0 and B_4^4 assuming that H is an external field of 5 mT and the magnetization M can be averaged according to $M = (M_c + 2M_a)/3$, where M_c and M_a are the magnetizations for fields along the c- and the a-axis respectively. The estimated molecular field constant was $-2.65 \cdot 10^6\ \text{mol/m}^3$ in the paramagnetic phase and $-2.98 \cdot 10^6\ \text{mol/m}^3$ in the ordered state. Acceptable fits to the experimental data have been obtained for sets of CEF parameters with B_2^0 between -8 and -9 K. The combination $B_2^0 = -8.78$ K, $B_4^0 = -0.054$ K and $B_4^4 = 2.79$ K has

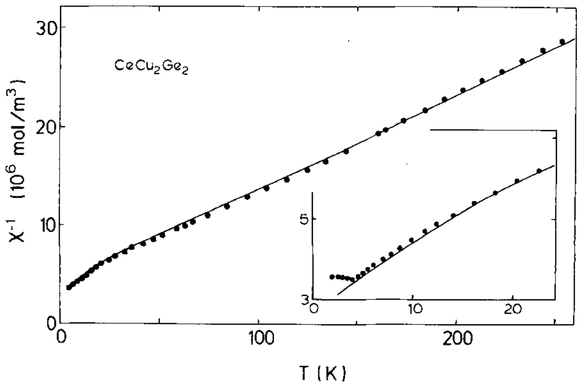

Fig. 6. Inverse dc susceptibility, χ^{-1} versus T, for $CeCu_2Ge_2$ (●) [7]. The lines represent the calculated paramagnetic susceptibility according to the level scheme of Fig. 8

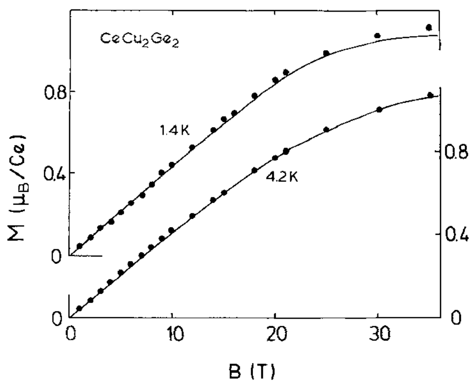

Fig. 7. High-field magnetization M as a function of magnetic field B for $CeCu_2Ge_2$ at 1.4 K and 4.2 K (●) [7]. The solid lines show results of a calculation (see text)

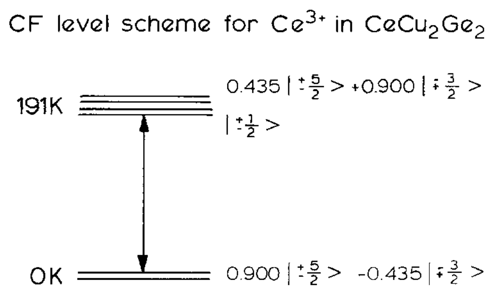

Fig. 8. Crystal-field level scheme of Ce^{3+} in the tetragonal compound $CeCu_2Ge_2$ as obtained from fits to the experimental results: $B_2^0 = -8.78$ K; $B_4^0 = -0.055$ K, $B_4^4 = 2.79$ K

been accepted as best solution. The temperature dependence of the paramagnetic susceptibility is shown in Fig. 6. The calculated magnetization as compared with experimental results is shown in Fig. 7. In both cases the agreement between observed and calculated data is satisfactory.

The corresponding level scheme of the Ce ion in the CEF is shown in Fig. 8. The ground state is predominantly $|5/2\rangle$. The easy axis of magnetization is along the c-direction.

A mean-field calculation has been carried out for the magnetically ordered phase. It was assumed that the molecular field is identical for all Ce ions and is directed along the easy c-axis. The calculated temperature dependence of the ordered moment is shown as a solid line in Fig. 3. It reaches a value of 1.54 μ_B at low temperatures, compared with the experimental value of 0.74 μ_B at 1.5 K. $CeCu_2Ge_2$ exhibits a reduction of the magnetic moment of the Ce ion by a factor of two. This seems to be a reasonable result taking the characteristic temperature scales into account: $T_K \approx 6$ K and $T_N \approx 4$ K. Obviously, the formation of a Kondo singlet state, accompanied by the cancellation of the Ce spin, has started. However, the antiferromagnetic intersite interactions are still strong enough to establish magnetic order at 4.1 K. The quenching of the Ce moments is documented in Fig. 3.

The energy splitting of the CEF ground doublet in the molecular field is practically proportional to the ordered moment. A value of 0.73 meV was calculated for $T=0$ K.

2. Magnetic relaxation rate. The width of the quasielastic line Γ (half width at half maximum) in the magnetic neutron scattering spectrum determines the magnetic relaxation rate. For the groundstate multiplet of a non-interacting local moment, the dynamic structure factor will exhibit a resolution limited peak at zero energy transfer. Interactions of the $4f$-moment with the conduction electrons introduce relaxation effects giving Lorentzian line shapes with an intrinsic width. In rare earth compounds, with a magnetically stable $4f$ configuration one expects a Korringa type of behavior for the quasielastic line width, namely $\Gamma = \alpha \cdot k_B T$. In rare earth ions with a well defined local moment the coefficient α is typically 10^{-3} [20]. Due to the strong coupling between the conduction electrons and the $4f$-electrons the magnetic relaxation rate in heavy fermion compounds behaves completely different: theoretical calculations [21–26] of the dynamic susceptibility show that $\Gamma(T)$ exhibits a finite line width for $T \to 0$ and deviates significantly from a linear T-dependence. Cox, Bickers and Wilkins [21] have performed a calculation of the magnetic relaxation rate for systems with non-stable $4f$-electron configurations in a wide temperature range ($0.01\,T_K \leqq T \leqq 40\,T_K$). They found that *i)* Γ is nonmonotonic with a minimum near the Kondo temperature and reaching a limiting low temperature value of $1.35\,T_K$. *ii)* the magnetic relaxation rate shows a non-linear high temperature behavior which is well fit by a square root law and *iii)* significant deviations from a Lorentzian line shape appear for $T \ll T_K$. However, this

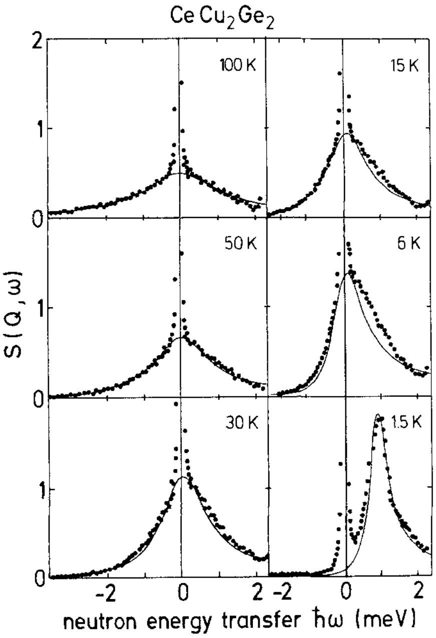

Fig. 9. Scattering law versus $\hbar\omega$ for an average scattering angle of $\Theta = 19$ deg. as obtained for $CeCu_2Ge_2$ at different temperatures ($T_N \cong 4.1$ K). The solid lines are the results of fits using a Lorentzian line

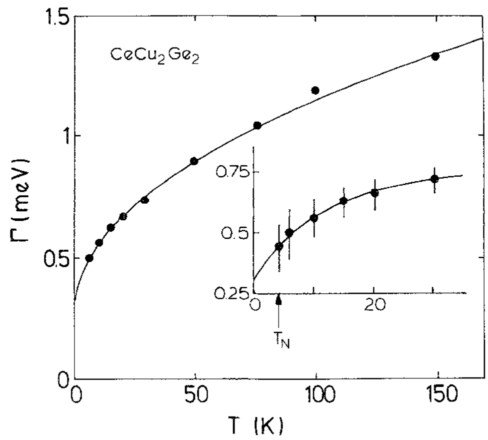

Fig. 10. Temperature dependence of the quasielastic line width (HWHM) for $CeCu_2Ge_2$. The solid lines were calculated using a square-root dependence of the magnetic relaxation rate

theory is restricted to ground states with a degeneracy $N_F \geqq 4$, and may not be strictly applicable in $CeCu_2Ge_2$ where the ground state is a doublet. Recently, magnetic relaxation rates in Kondo systems have been calculated taking CEF effects into account [22]. Kuramato and Müller-Hartmann [23] derived an analytic result for the dynamic susceptibility within the degenerate Anderson model. For $N_F > 2$ they found an increasingly asymmetric line shape with an increasing degeneracy of the ground state. However, this analytic result is not valid for $\omega \to 0$.

The measurements of the quasielastic line widths have been performed at the time-of-flight spectrometer IN6 located at the cold source of the High Flux Reactor at the *ILL*. Incident neutron energies of 3.15 meV have been used. Some of the data as obtained for $CeCu_2Ge_2$ are shown in Fig. 9. The half widths at half maximum (HWHM) of the quasielastically scattered neutrons are a direct measure of the magnetic relaxation rate. Already from a first inspection it is clear that the magnetic relaxation rates are strongly enhanced when compared to rare earth systems with stable moments [20]. At 1.5 K, which is well below T_N, a well defined excitation at approx. 1 meV is clearly observable, which we interpret as a density of magnon states in the magnetically ordered phase. In the paramagnetic state ($T \geqq 4.1$ K) the quasielastic lines can, in a first approximation, be described by a Lorentzian line shape weighted with a detailed balance factor. The solid lines in Fig. 9 show the results of the best fits to the experimental data. For $T \leqq 30$ K there appear significant deviations from a Lorentzian line shape. These deviations point towards short range order correlations. Figure 10 shows the temperature dependence of the half width at half maximum as determined from the fits to $S(Q, \omega)$. The temperature dependence of the HWHM directly determines the magnetic relaxation rate $\Gamma(T)$. In $CeCu_2Ge_2$ it can be described well using

$$\Gamma(T) = \Gamma_0 + \gamma\sqrt{T}$$

with a residual line width $\Gamma_0 = 3.5$ K and a coefficient $\gamma = 0.98\,(K^{1/2})$. From thermodynamic and transport properties we have characterized $CeCu_2Ge_2$ as a Kondo lattice with a Kondo temperature $T_K = \cong 6$ K [7, 14, 18]. According to Cox et al. [21] one would expect a minimum in $\Gamma(T)$ at T_K and a square root T-dependence for higher temperatures. No indications of a minimum at 6 K are detectable in Fig. 9. However, the magnetic ordering temperature is close to T_K and the Kondo minimum as well as the limiting low temperature value $\Gamma(T \to 0) = 1.35\,T_K$ could be screened by the onset of magnetic order. In the next chapter we will see that a refined analysis, taking critical spin fluctuations into account, yields a minimum near $T \approx 10$ K.

The Fermi-liquid description makes some specific predictions for the imaginary part of the magnetic susceptibility (2). E.g. within the framework of a Fermi-liquid theory a linear dependence of the magnetic relaxation rate on the momentum transfer Q is expected [27]. We observed that in $CeCu_2Ge_2$ Γ is independent of Q over a wide range of momentum transfers (0.4 $\text{Å}^{-1} < Q < 2.3$ Å^{-1}).

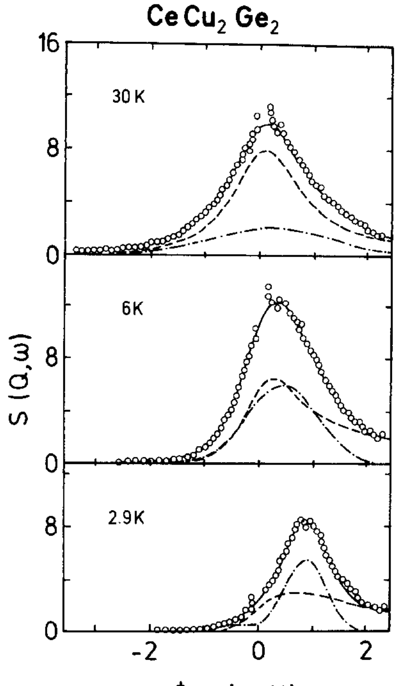

Fig. 11. Scattering law versus $\hbar\omega$ in $CeCu_2Ge_2$ for temperatures $T \lesssim T_N$. The solid lines represent the results of fits using a superposition of a Lorentzian (dashed lines) and a Gaussian contribution (dashed-dotted lines)

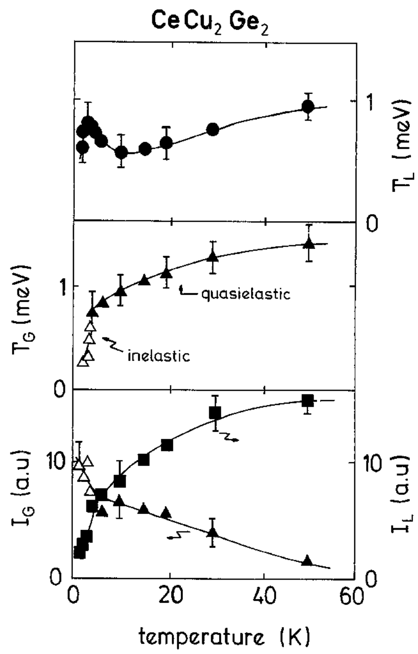

Fig. 12. Temperature dependence of the intensities and widths of the Lorentzian and Gaussian components. The lines are drawn to guide the eye. *Upper frame:* quasielastic Lorentzian width (●) (Γ_G), *middle frame:* quasielastic (▲) and inelastic (△) Gaussian width (Γ_G), *lower frame:* elastic (▲) and inelastic (△) Gaussian intensities (I_G); quasielastic Lorentzian intensities (I_L) (■)

3. Critical spin fluctuations and excitations in the magnetically ordered phase. Figure 9 demonstrates that significant deviations from a Lorentzian line shape appear for temperatures close to T_N ($30\,K \leqq T \leqq T_N = 4.1$ K). Two possible explanations can be given:

i) The extra contributions are due to inelastic excitations within the paramagnetic state (paramagnons) indicating that the degeneracy of the ground state doublet is partially removed or

ii) the deviations from the Lorentzian line shape are associated with critical spin fluctuations which can be approximately described by quasielastic Gaussian lines [28]. Critical spin fluctuations can occur as precursors of the magnetic ordering process and have been observed in a number of magnetically ordered heavy-fermion systems [28].

In Fig. 11 we demonstrate, that for $T \geqq T_N$ these extra intensities can be described with a quasielastic Gaussian component, and hence indicate the existence of spin fluctuations. Below the Néel temperature a superposition of an inelastic Gaussian and a quasielastic Lorentzian component had to be used. The former describes magnetic excitations, the latter is a measure of the $4f - l$ conduction electron-hybridization. This result shows, that magnetic relaxation still exists in the magnetically ordered state, even well below the ordering temperature, in agreement with the idea of a reduced ordered moment. The quasielastic component would be absent in a "normal" magnetic system. Hence we suggest that the persistence of the quasielastic component is characteristic for HFS and corresponds to the enhanced linear term in the specific heat for $T < T_N$ (see Fig. 1 and [7]).

Figure 12 shows a summary of the results for the temperature range where a superposition of Gaussian and Lorentzian contributions gave a considerable improvement of the fit ($T \leqq 50$ K). In the upper frame the quasielastic Lorentzian width Γ_L is plotted versus temperature. Using this improved analysis Γ_L passes through a minimum near 10 K. According to the theory by Cox et al. [21] this minimum defines the characteristic temperature of the system $T_K = 10$ K which is not too different from that as obtained from estimates using thermodynamic and transport data. Below T_N, Γ_L slightly decreases. We are aware that on the basis of the present data it is hard to prove the existence of a minimum in $\Gamma(T)$. A constant finite line width for $T \to 0$ is still within the experimental uncertainties. It would be highly desirable to study non-ordering HFS at low T [29].

The middle frame of Fig. 12 shows the temperature dependence of the Gaussian width $\Gamma_G(T)$ which describes the antiferromagnetic intersite correlations. For $T > T_N$ $\Gamma_G(T)$ slightly decreases with decreasing

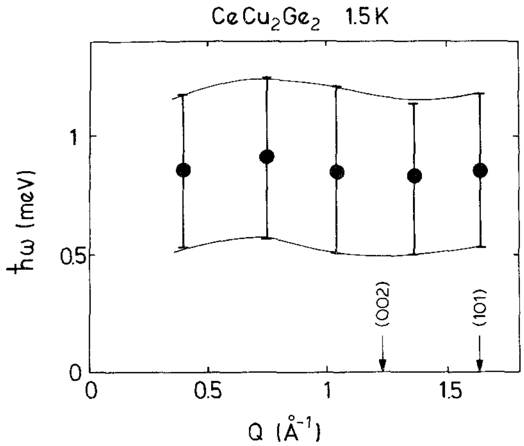

Fig. 13. Momentum transfer dependence of the width of the magnetic excitations in $CeCu_2Ge_2$ at 1.5 K. The bars represent the full width at half maximum of the inelastic contributions. The positions of the first two nuclear Bragg peaks are indicated

temperatures indicating an increasing lifetime of critical spin fluctuations. Below T_N the width abruptly decreases. Now the Gaussian components are shifted towards finite frequencies. They describe the magnetic excitations of $CeCu_2Ge_2$. For ordinary Heisenberg antiferromagnets one would expect propagating spin waves with a linear dispersion relation for small magnon wave vectors q and furthermore that the magnon dispersion (band width) increases with decreasing temperatures, in addition to an overall splitting of the ground state doublet.

In the lower frame of Fig. 12 the temperature dependence of the relative intensities of Gaussian and Lorentzian components are given. Below T_N the magnetic excitations carry the major part of the scattered intensity. But the main results is that the quasielastic component persists down to the lowest temperatures, which presumably is related to the Kondo-derived reduction of the ordered Ce-moment and to the enhanced linear term γT in the specific heat. Similar conclusions have been drawn for the archetypical system $CeAl_2$ [30]. In contrast to $CeCu_2Ge_2$, however, γ appears to be only weakly temperature dependent in $CeAl_2$.

The splitting of the ground state develops gradually in the ordered phase. The size of the splitting $\hbar\omega = 0.85$ meV, is compatible with the magnetic moment of the ground state. The line width is decreasing with temperature which we interpret that dispersion effects due to exchange interactions do not play an important role. Figure 13 shows the Q-dependence of the magnetic excitations. Again we find that the overall line shapes are dominated by the mean-field splitting of the ground state. Of course, one has to bear in mind that soft modes might have a low weight in these zone-averaged data.

III. Conclusions

$CeCu_2Ge_2$ is a well characterized Kondo lattice with a magnetically ordered ground state. Anomalous transport and thermodynamic properties have been reported. Resistivity [12], thermoelectric power [14] and specific heat data [17] point towards a Kondo temperature $T_K \cong 6$ K. In addition an enhanced linear term in the specific heat can still be found at low T well below the magnetic phase transition.

The results of this elastic and inelastic neutron scattering study can be summarized as follows:

– at $T_N = 4.1$ K an incommensurate magnetic order develops. The spin structure can be described by a spiral with an ordering wave vector $\mathbf{q}_0 = (0.28, 0.28, 0.54)$ and with a plane of rotation perpendicular to the propagation vector. The ordered moment was found to be $\mu_S = 0.74\mu_B$ at 1.5 K.

– only one inelastic transition due to crystal electric field effects could be detected at $\hbar\omega/k_B = 191$ K. A determination of the crystal electric field parameters of the Ce ion in a tetragonal point symmetry was possible utilizing complementary susceptibility and magnetization data. The wave function of the ground state doublet was found to be $0.9|\pm 5/2\rangle - 0.435|\pm 3/2\rangle$ yielding a magnetic moment of $1.54\mu_B$. Thus, due to the formation of a Kondo singlet, $CeCu_2Ge_2$ exhibits a large reduction of the magnetic moment of the Ce ion.

We performed a detailed quasielastic neutron scattering study on the line width of the ground state in the paramagnetic state. An anomalous temperature dependence of the magnetic relaxation rate was detected. $\Gamma(T)$ is characterized for an unstable $4f$-ion and pointed towards a Kondo temperature $T_K \cong 10$ K. For temperatures close to the antiferromagnetic ordering transition deviations from the Lorentzian line shape were detected which we ascribe to critical spin fluctuations.

– in the magnetically ordered state we observed, in addition to a broad band of magnetic excitations at 0.85 meV, a quasielastic component, even well below T_N. This quasielastic line seems to be characteristic for heavy-fermion systems and corresponds to an enhanced value of the linear term of the specific heat.

This work was supported by the Bundesministerium für Forschung und Technologie under the contract number 03-L01MAI-0 and by the Sonderforschungsbereich 252, Darmstadt/Frankfurt/Mainz/Stuttgart.

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